

PATENT SPECIFICATION

(11) 1320 169

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NO DRAWINGS

- (21) Application No. 29213/70 (22) Filed 16 June 1970
 (31) Convention Application No. 833806 (32) Filed 16 June 1969 in
 (33) United States of America (US)
 (44) Complete Specification published 13 June 1973
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C2P 1L1 3B12B 3B15B 3B18C 3B19A 3B19B 3B19C
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(54) PHOSPHITE ESTERS OF HINDERED PHENOLS

(71) We, ASHLAND OIL, INC., a Corporation organised and existing under the laws of the State of Kentucky, United States of America, of 1409 Winchester Avenue, Ashland, Kentucky 41101, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to phosphite esters. More particularly, it relates to reaction products of phosphite-ester-forming phosphorus compounds and hindered bis phenols.

Synthetic polymers such as polyethylene, rubber, waxes, oils, fats and numerous other compounds are attacked by oxygen and eventually may become useless for their intended purpose. The chemical reaction by means of which oxygen attacks and degrades the compounds is a free radical chain reaction. Free radicals are produced by abstraction of hydrogen atoms from the molecules of the compounds by, for example, light, heat, mechanical action or active molecules.

The free radicals are extremely reactive in the presence of air and oxygen, forming peroxide radicals, which, in turn, abstract hydrogen atoms from the molecules to form additional radicals. These again react with oxygen in the same manner. Thus, once started, the reaction is a self-perpetuating, degradative, continuous chain reaction until stopped. In order to prevent such degradation, various antioxidants have been added which react with and destroy the intermediate chemical free radicals as they form, without producing equally reactive intermediates.

Various phosphorus containing compounds and various phenols such as 2,2'-methylenebis- (6-tert.butyl-4-methylphenol) have been used as antioxidants; however, the prior antioxidants have the disadvantage of losing effectiveness at comparatively high temperatures, even when used in synergistic combinations. The invention has therefore been made with this point in mind.

According to the invention there is provided a phosphite ester in which at least one of the available bonds on the phosphorus atom is connected to a para-substituted hindered (as hereinafter defined) phenol through the oxygen remaining after removal of the hydrogen of a phenolic hydroxyl group, the para-substituted hindered phenol being a hindered bisphenol with the phenolic rings bridged through a saturated aliphatic hydrocarbon linkage of 1 to 8 carbon atoms, and with a hydroxyl group on each ring being ortho and/or para to the linkage, and with both of the positions ortho to the hydroxyl group on each ring being substituted with a hydrocarbon, and in which all of the available bonds on the phosphorus atom not connected to a para-substituted hindered phenol are connected through the oxygen remaining after removal of the

PATENTS ACT 1949

SPECIFICATION NO 1320169

Reference has been directed, in pursuance of Section 9, subsection (1) of the Patents Act 1949, to Specification No 1108333.

THE PATENT OFFICE
 26 July 1973

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The free radicals are extremely reactive in the presence of air and oxygen, forming peroxide radicals, which, in turn, abstract hydrogen atoms from the molecules to form additional radicals. These again react with oxygen in the same manner. Thus, once started, the reaction is a self-perpetuating, degradative, continuous chain reaction until stopped. In order to prevent such degradation, various antioxidants have been added which react with and destroy the intermediate chemical free radicals as they form, without producing equally reactive intermediates.

Various phosphorus containing compounds and various phenols such as 2,2'-methylenebis-(6-tert-butyl-4-methylphenol) have been used as antioxidants; however, the prior antioxidants have the disadvantage of losing effectiveness at comparatively high temperatures, even when used in synergistic combinations. The invention has therefore been made with this point in mind.

According to the invention there is provided a phosphite ester in which at least one of the available bonds on the phosphorus atom is connected to a para-substituted hindered (as hereinafter defined) phenol through the oxygen remaining after removal of the hydrogen of a phenolic hydroxyl group, the para-substituted hindered phenol being a hindered bisphenol with the phenolic rings bridged through a saturated aliphatic hydrocarbon linkage of 1 to 8 carbon atoms, and with a hydroxyl group on each ring being ortho and/or para to the linkage, and with both of the positions ortho to the hydroxyl group on each ring being substituted with a hydrocarbon, and in which all of the available bonds on the phosphorus atom not connected to a para-substituted hindered phenol are connected through the oxygen remaining after removal of the hydrogen of the phenolic hydroxyl groups to a hindered mono or poly phenolic compound which is a hydroxy substituted phenol or a phenol with at least two of the positions ortho and para to the hydroxy group on each phenol ring substituted.

It has been found that the compounds of the invention surprisingly and unexpectedly exhibit increased stabilizing properties; and thereby the quantity necessary to achieve a desired level of stabilization at such temperatures as are normally used for moulding, calendaring, extrusion and other forming processes is reduced. Moreover, with the compounds of the invention, substantial degrees of stabilization can be attained at those higher temperatures at which combinations of phosphorus compounds separate and bis phenolic compounds are ineffective.

[Price 25p]

The compounds of this invention are phosphite esters in which at least one of the available bonds on the phosphorus atom is connected to a para-substituted hindered phenol through the oxygen remaining after the removal of the hydrogen of a phenolic hydroxy group. The para-substituted hindered phenol is a hindered bisphenol with the two phenolic rings bridged through a saturated aliphatic hydrocarbon linkage of 1 to 8 carbon atoms, and with one or two hydroxyl groups on each ring being ortho or para to the saturated aliphatic hydrocarbon linkage, and with both of the positions ortho to the OH group or groups on each ring being substituted with hydrocarbon groups of at least one carbon atom. The term 'para-substituted hindered' means that all positions ortho and para to the OH group on each ring of the hindered bisphenol are substituted. All available bonds on the phosphorus atom not connected to the para-substituted hindered phenol are connected to a hindered phenolic compound through the oxygen remaining after removal of the hydrogen of the phenolic hydroxy group. Preferably the hydroxyl group on at least one ring of the para substituted hindered phenol is ortho to the saturated aliphatic hydrocarbon linkage.

The compounds of the invention may constitute, for example, the reaction products of one reacted mole of a phosphite ester-forming phosphorus compound and at least one reacted mole and preferably three reacted moles of a para substituted hindered phenol (as defined above) and if necessary, at least one other hindered phenolic compound in an amount so that each of the available bonds on the phosphorus atom is connected to a hindered phenol through the oxygen remaining after the removal of the hydrogen of a phenolic hydroxyl group.

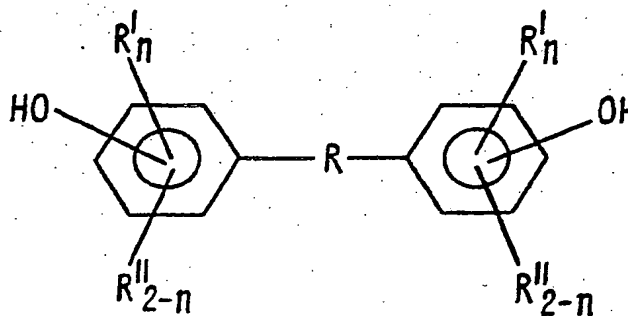
The reaction products are formed under known phosphite ester-forming conditions of time and temperature with known catalysts.

The production of the novel compounds of our invention was unexpected since it was generally believed heretofore among those of ordinary skill in the art that the hydroxyl groups in fully hindered phenols such as one used herein, are virtually unreactive and cannot be converted to various derivatives by normal techniques. For example, this low reactivity of hindered phenols is discussed in Kirk and Othmer, *Encyclopedia of Chemical Technology*, Vol. 1, pages 902 and 906. Indeed, it is quite surprising and unexpected that the hindered phenols used herein react with the phosphorus compounds under normal phosphite ester-forming conditions.

The phosphorus compounds used in preparing the compounds of our invention can be any of the known phosphite ester-forming compounds. Examples of such compounds are the phosphorus trihalides, such as the chloride, bromide, and iodide. The most preferred phosphorus compound is phosphorous trichloride.

The para-substituted hindered phenol compounds are represented by the following formula:

I.



R is a saturated aliphatic hydrocarbon linkage or group (alkylidenes and alkylenes) having 1—8 carbon atoms, and preferably having 1—4 carbon atoms. More preferably, R can be methylene, ethylene, isopropylene, ethylidene, n-propylidene, and isopropylidene. The most preferred R group is methylene.

The OH group on each ring is either ortho or para to the hydrocarbon linkage and is most preferably in the ortho position.

Each R' individually is a hydrocarbon group of at least one carbon atom and is ortho to the OH group on each ring. Usually the hydrocarbon group is free of non-benzenoid unsaturation. R' is preferably a hydrocarbon group of from 1—22 carbon

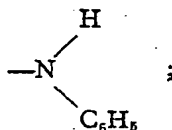
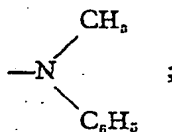
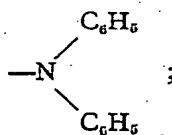
atoms and preferably an alkyl group such as methyl, ethyl, t-butyl, t-amyl, t-hexyl, cyclohexyl, t-pentyl, t-octyl, phenyl, naphthyl, α methylcyclohexyl, nonyl, benzyl, methyl, isobornyl, anthranyl, phenanthranyl, norbornyl, cyclopropyl, cyclopentyl, bicyclohexyl, cyclobutyl, 1,2-dimethylcyclopropyl, and xylyl, more preferably R' is a hydrocarbon of from 1—12 carbon atoms, and even more preferably is an alkyl group containing 1—12 carbon atoms of which t-butyl is the most preferred.

n=1 or 2.

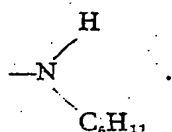
R'' is any substituent which will occupy the position para to the OH group on each ring, when it is unfilled, in order to prevent the reactants from combining through said para position. R'' advantageously is, but is not limited to, a hydrocarbon group, such as the hydrocarbon groups set forth above for R', or a halogen atom such as chlorine or bromine; or —NO₂; or —SR'''; or —OR'''; or —COOR'''; or —NR''R'''; or —NHR^{IV}NH₂; or —NHOH; —NHR^{IV}OH; wherein R'' is a hydrocarbon group as defined above for R', and R^{IV} is an alkylene group of 1—22 carbon atoms and preferably of 1—12 carbon atoms. Such specific —SR''' groups are —SH; —SCH₃; —SC₂H₅; —SC₃H₇; —SC₄H₉; and —SC₆H₁₁. Some specific —OR''' groups are —OC₆H₅; —OC₈H₁₁; —OCH₃; —OC₂H₅; and —OC₆H₁₁.

Some specific —COOR''' groups are —COOH; —COOC₂H₅; —COOC₆H₁₁; —COOC₈H₁₇; —COOC₁₀H₂₁; and —COOCH₃.

Some specific —NR''R''' groups are —NH₂; —NHCH₃; —N(CH₃)₂;



—NHC₆H₁₁; —N(C₆H₁₁)₂; —N(C₆H₁₁)₂; and



Some specific —NHR^{IV}NH₂ groups are —NHCH₂NH₂; —NHC₂H₄NH₂; —NHC₆H₁₁NH₂; and —NHC₆H₁₂NH₂.

Some specific —NHR^{IV}OH groups are —NHCH₂OH; —NHC₂H₄OH; —NHC₆H₁₁OH; and —NHC₆H₁₂OH.

R'' preferably is an alkyl group with methyl being the most preferred. Usually alkyl groups of not more than 22 carbon atoms are employed. Advantageously, the alkyl group contains 1—12 carbon atoms.

Specific examples of suitable bisphenols are 4,4'-methylenebis-(2,6-dimethylphenol); 2,2' - methylenebis - (4 - ethyl - 6 - tert. - butylphenol); 2,2' - methylenebis - (4 - chloro - 6 - tert. - butylphenol); 2,2' - methylenebis - (4,6 - ditertiarybutylphenol); 2,2' - methylenebis - (4,6 - dimethylphenol); 2,2' - isopropylidene bis - (4,6 - ditertiarybutylphenol); 2,2' - methylenebis - (6 - nonyl - 4 - methylphenol); 2,2' - ethylidene bis - (4,6 - ditertiarybutylphenol); and 4,4' - methylenebis - (2,6 - ditertiarybutylphenol). The preferred para-substituted hindered phenol is 2,2' - methylenebis - (4 - methyl - 6 - tertiarybutylphenol).

The other hindered phenol is a mono- or polyphenolic compound which may be either a hydroxy substituted phenol, or a phenol with at least two of the positions ortho and para to the hydroxy group on each phenolic ring being substituted with the appro-

priate ortho and/or para substituents as defined above for the para-substituted hindered phenol.

Suitable examples of the hydroxy substituted phenol are catechol, resorcinol, and hydroquinone. Suitable examples of a phenol with at least two of the positions ortho and para to the hydroxy group on each phenolic ring being substituted are 2-tertiary-butyl-4-methylphenol; 2 - tertiarybutyl - 4 - chlorophenol; 2,6 - dimethylphenol; 2,6 - ditertiarybutyl - 4 - methylphenol; 4,4 - bis - (2,6 - ditertiarybutylphenol); 2,4,6 - trimethylphenol; and 2-methyl-4-n-butylphenol.

The catalyst used in preparing the phosphorus compounds of our invention can be any of the conventional catalysts used in preparing phosphite esters. Examples of suitable catalysts are metals such as copper powder, iron filings, calcium, aluminum, and magnesium; halides such as aluminum chloride, magnesium chloride, ferric chloride, tin tetrachloride, zinc chloride, boron trifluoride, titanium trichloride, and titanium tetrachloride; sulfates such as copper sulfate; and oxides such as magnesium oxide and copper oxide. The preferred catalyst is magnesium. The catalyst is usually present in amounts between 0.5% and 5% by weight, based on the weight of the phenolic compounds.

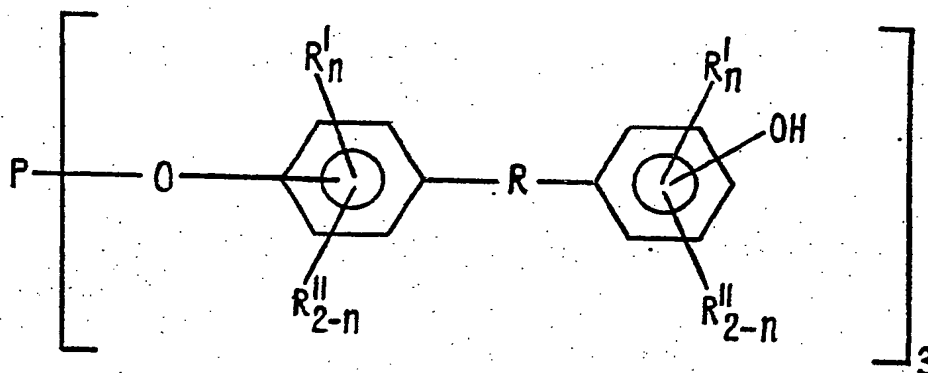
The diluent can be any liquid, provided it is inert (not reactive in any manner which will harm the reaction or the product), and should dissolve at least one of the reactants. Examples of suitable solvents are the chlorinated hydrocarbons. The preferred solvent is o-dichlorobenzene. The diluent is most commonly present in amounts between 5% and 300% by weight, based on the weight of the phenolic compounds.

The process is not limited to specific reaction temperatures, since the reaction can be carried out over a wide range of temperatures. For example, the process can be carried out at temperatures of from about 0°C to temperatures of about 200°C. The preferred temperature range varies from about 150°C to 200°C, and the most preferred temperature range is from about 160°C to 170°C. Also the process is not limited to any specific reaction time, since the time required will vary, primarily dependent upon the particular reactants, temperatures, and catalysts. Preferably, the reaction times vary from about 12 to about 72 hours. About 24 hours is the reaction time which is most commonly used.

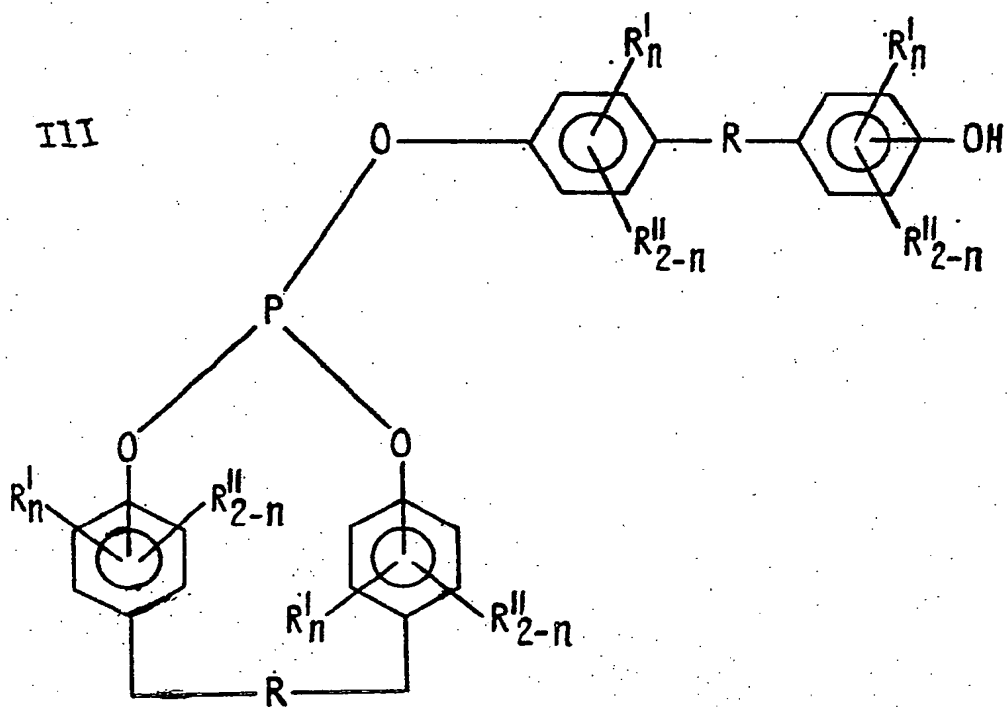
Atmospheric pressure is the most commonly used pressure for carrying out the invention. Of course, higher or lower pressures can be employed when desired.

Some of the novel phosphite esters of the invention are illustrated by the following structures:

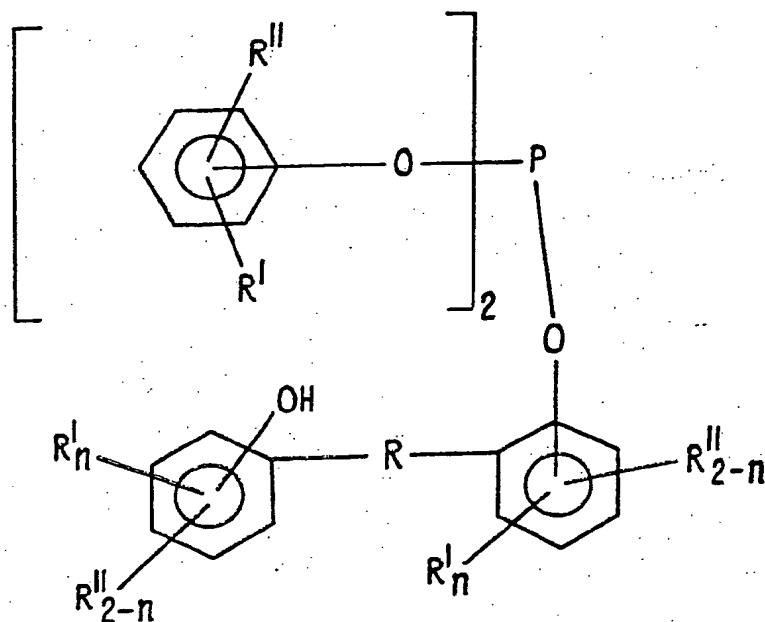
II



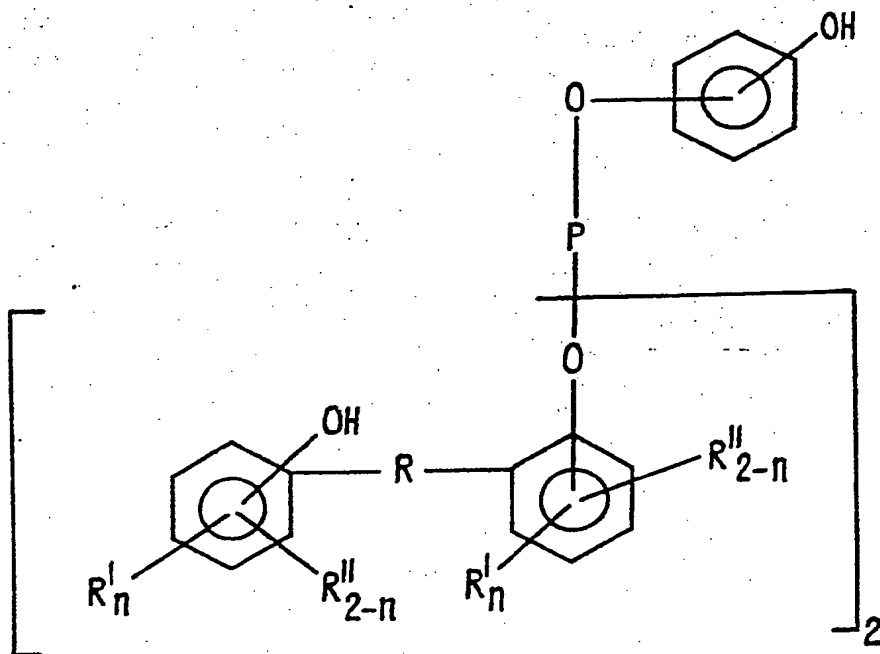
III



IV.



v.



wherein R, R', and R'' and n have the same definitions and positions set forth in respect to Formula I.

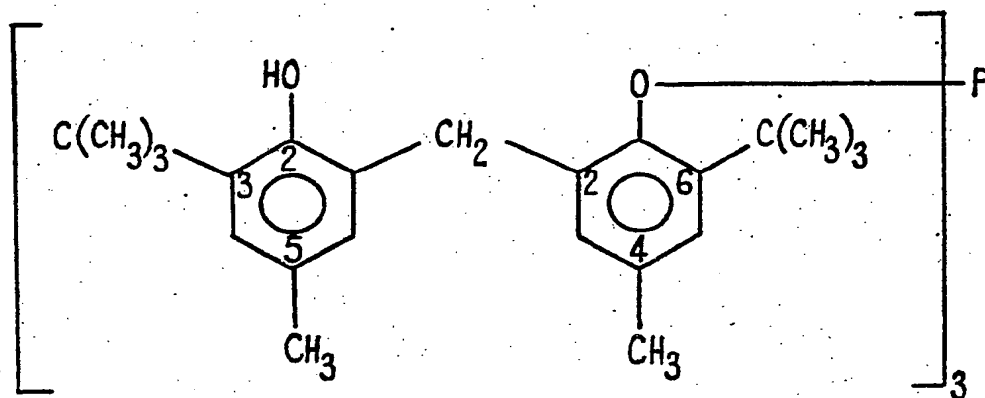
The phosphorus compounds of the invention are useful as antioxidants in a wide variety of materials. Among such materials are synthetic polymers, rubber, waxes, fats and oils. Among the synthetic polymers which can be stabilized with the products of this invention are: polyolefins such as polyethylene, polypropylene and polybutene;

diene rubbers such as polyisoprene, polybutadiene, copolymers of conjugated dienes and at least one other copolymerizable monomer such as styrene, acrylonitrile, methyl acrylate, and 2-vinyl pyridine, polystyrene; polyacrylates; polyester; epoxy resins; polyacetals; vinyl chloride polymers and polyurethanes. The products of this invention are particularly effective in stabilizing polyolefins, e.g. polypropylene and polyethylene. The amount of antioxidant needed to stabilize a particular amount of polymeric material can obviously be varied over a wide range of proportions dependent upon the identity of the specific polymeric material, the desired degree of stabilization, and the environment in which the material is to be used. An amount of about 0.1% by weight based upon the antioxidant and the material to be stabilized is very effective. The threshold at which the compounds of the invention are effective is about 0.001% by weight based upon the weight of material to be stabilized. The compounds of the invention are used in amounts as high as 5% by weight or higher based upon the weight of the material to be stabilized.

The compounds of this invention have exhibited performance superior to the prior art antioxidants to which they have been compared as will be illustrated in the following examples, in which all parts are weight unless the contrary is stated.

Example 1

To a mixture of 3 moles of 2,2'-methylenebis (4-methyl-6-tertiarybutylphenol), 3900 parts of o-dichlorobenzene, as an inert solvent, and about 4 moles of a magnesium metal catalyst in a glass-lined container or flask equipped with a glass-surfaced stirrer or agitator is added all at once and with agitation, 1 mole of phosphorus trichloride. The resultant mixture is agitated for 24 hours at a pressure of about one atmosphere with the temperature being maintained between 160° and 170°C. Hydrogen chloride gas evolves as the reaction proceeds and is neutralized in a scrubbing device. The reaction mixture is cooled, 1000 parts of water are added and the magnesium catalyst is separated by filtration. The clarified mixture is then neutralized to a pH between 6 and 8 by use of an aqueous sodium carbonate solution and after washing with 1000 parts of water, the organic portion is then separated from the aqueous portion of the mixture. The reaction product and inert solvent are then separated by vacuum distillation. The reaction product is a clear, friable glass melting between 59° and 64°C., and has the following structure as determined by elemental analysis and IR spectra:

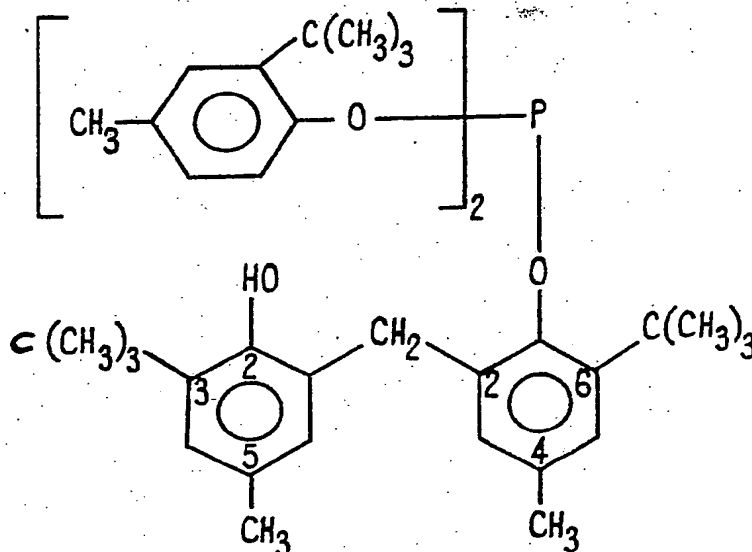


tris [2 - (2 - hydroxy - 3 - t - butyl - 5 - methylbenzyl) - 4 - methyl - 6 - t - butylphenyl] Phosphite

Example 2

To a mixture of 2 moles of 2-tertiarybutyl-4-methylphenol, 1 mole of 2,2'-methylenebis (4-methyl-6-tertiarybutylphenol), 3900 parts of o-dichlorobenzene, as an inert solvent, and .4 moles of a magnesium metal catalyst in a glass-lined container or flask equipped with a glass-surfaced stirrer or agitator is added all at once and with agitation, 1 mole of phosphorus trichloride. The resultant mixture is agitated for 24 hours at a pressure of about one atmosphere with the temperature being maintained between 160° and 170°C. Hydrogen chloride gas evolves as the reaction proceeds and is neutralized in a scrubbing device. The reaction mixture is cooled, 1000 parts of water are added and the magnesium catalyst is separated by filtration. The clarified

mixture is then neutralized to a pH between 6 and 8 by use of an aqueous sodium carbonate solution. The organic portion of the mixture is then washed with 1000 parts of water, and an organic layer is then separated from the aqueous portion of the mixture. The reaction product and inert solvent are then separated by vacuum distillation. The reaction product, di[(2 - tertiarybutyl - 4 - methylphenyl), mono(2 - (2 - hydroxy - 3 - tertiarybutyl - 5 - methylbenzyl) - 4 - methyl - 6 - tertiarybutylphenyl]phosphite, has the following structure as determined by elemental analysis and IR spectra:



Example A

The product of Example 1 is admixed with a polyethylene of 0.960 specific gravity and approximately 300,000 molecular weight in a steel container and the mixture is extruded twice at 300°F. The concentration of the product of Example 1 is then adjusted to 0.1% by weight of the polymer by the addition of more of the polyethylene and the mixture is again extruded twice at 300°F. The resulting polyethylene composition is then pressed into a 6—6.5 mil film at 310°F and 1280 psi on a 10" hydraulic ram press. Likewise, a film of the same polyethylene without any antioxidant and a film of the same polyethylene containing 0.1% by weight of some other antioxidants are prepared by the method set forth above. The resulting films are then subjected to 125° ± 1°C. in a forced draft oven. The absorbance in the carboxyl region of the IR Spectrum (5.8 Angstroms) is then recorded after periods of exposure. When the observed absorbance reaches 94%, the sample is considered to be "oxidized", and the time of exposure to reach this point is recorded in the table below:

Additive	Time in Hours to reach 94% Amount Absorbance	
None		50
2,2'Methylenebis (6 - t - butyl - 4 - methylphenol)	0.1%	125
Reaction product of Example 1	0.1%	320
Reaction product of Example 2	0.1%	173

Additive	Time in Hours to reach 94% Amount Absorbance	
	0.1%	60
	0.1%	60
	0.1%	74
	0.10%	90

From the above table, it is readily apparent that the phosphite esters of the invention exhibit greatly improved stabilizing properties as compared to stabilizers outside the scope of the invention.

WHAT WE CLAIM IS:—

1. A phosphite ester in which at least one of the available bonds on the phosphorus atom is connected to a para-substituted hindered (as herein defined) phenol through the oxygen remaining after removal of the hydrogen of a phenolic hydroxyl.

- group, the para-substituted hindered phenol being a hindered bisphenol with the phenolic rings bridged through a saturated aliphatic hydrocarbon linkage of 1 to 8 carbon atoms, and with a hydroxyl group on each ring being ortho or para to the linkage, and with both of the positions ortho to the hydroxyl group on each ring being substituted with a hydrocarbon, and in which all of the available bonds on the phosphorus atom not connected to a para-substituted hindered phenol are connected through the oxygen remaining after removal of the hydrogen of the phenolic hydroxyl groups to a hindered mono or poly phenolic compound which is a hydroxy substituted phenol or a phenol with at least two of the positions ortho and para to the hydroxyl group on each phenol ring substituted.
- 5 2. A phosphite ester as claimed in claim 1 in which the saturated aliphatic hydrocarbon linkage of the para-substituted hindered phenol contains from 1 to 4 carbon atoms.
- 10 3. A phosphite ester as claimed in claim 2 in which the saturated aliphatic hydrocarbon linkage of the para substituted hindered phenol is a methylene linkage.
- 15 4. A phosphite ester as claimed in any preceding claim in which the hydrocarbon substituent of the para substituted hindered phenol contains from 1 to 22 carbon atoms.
5. A phosphite ester as claimed in claim 4 in which the hydrocarbon substituent of the para substituted hindered phenol is an alkyl group.
- 20 6. A phosphite ester as claimed in claim 5 in which the hydrocarbon substituent is an alkyl group containing from 1 to 12 carbon atoms.
7. A phosphite ester as claimed in Claim 6 in which the hydrocarbon substituent is a tert-butyl group.
- 25 8. A phosphite ester as claimed in any preceding claim in which the hydroxyl group on at least one ring of the para-substituted hindered phenol is ortho to the saturated aliphatic hydrocarbon linkage.
9. A phosphite ester as claimed in Claim 1 in which the para-substituted hindered phenol is 2,2' - methylenebis - (4 - methyl - 6 - tert - butylphenol).
- 30 10. A phosphite ester as claimed in any preceding claim in which the position para to the OH group on each ring of the para substituted hindered phenol is substituted with a hydrocarbon group, a halogen atom, $-\text{NO}_2$, $-\text{SR}'''$, $-\text{OR}'''$, $-\text{COOR}'''$, $-\text{NR}''' \text{R}'''$, $-\text{NHR}^{\text{IV}} \text{NH}_2$, $-\text{NHOH}$, and $-\text{NHR}^{\text{IV}} \text{OH}$, where R''' represents a hydrocarbon group having from 1 to 22 carbon atoms, and R^{IV} represents an alkylene group having from 1 to 22 carbon atoms.
- 35 11. A phosphite ester as claimed in Claim 1 in which the position para to the OH group on each ring of the hindered bis phenol is substituted with an alkyl group having from 1 to 22 carbon atoms.
12. A phosphite ester as claimed in Claim 11 in which the alkyl group is a methyl group.
- 40 13. A phosphite ester as claimed in any preceding claim in which all three of the available bonds on the phosphorus atom each are connected to a para-substituted hindered phenol as defined in any preceding claim through the oxygen after removal of the hydrogen of a phenolic hydroxy group.
- 45 14. Tris - (2 - (2 - hydroxy - 3 - tert - butyl - 5 - methylbenzyl) - 4 - methyl - 6 - tert - butylphenyl) phosphite.
15. Di - (2 - tert - butyl - 4 - methylphenyl), mono(2 - (2 - hydroxy - 3 - tert - butyl - 5 - methylbenzyl)4 - methyl - 6 - tert - butylphenyl) phosphite.
- 50 16. The reaction product of one reacted mole of a phosphite ester-forming phosphorus compound and at least one reacted mole of a para-substituted hindered (as herein defined) phenol, and optionally a hindered phenolic compound which is a hydroxy substituted phenol or a phenol with at least two of the positions ortho and para to the hydroxyl group on the phenol ring substituted in an amount so that all of the available bonds on the phosphorus atom are connected to a hindered phenol through the oxygen remaining after removal of the hydrogen of a phenolic hydroxyl group; in which the para-substituted hindered phenol is a hindered bisphenol with the phenolic rings bridged through a saturated aliphatic hydrocarbon linkage of 1 to 8 carbon atoms,
- 55

and with one hydroxyl group on each ring being ortho or para to the saturated aliphatic hydrocarbon linkage, and with both of the positions ortho to the OH group on each ring substituted with a hydrocarbon group, and with all positions ortho and para to the OH group on each ring substituted.

5 17. The reaction product claimed in Claim 16 in which all three of the available bonds on the phosphorus atom each are connected to the para-substituted hindered phenol through the oxygen remaining after removal of the hydrogen of a phenolic hydroxyl group.

5

18. A phosphite ester according to Claim 1 and substantially as herein described.

For the Applicants:
LLOYD WISE, BOULY & HAIG,
Chartered Patent Agents,
Norman House,
105—109 Strand,
London, WC2R 0AE.

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